

# Bulk Metallic Glasses: At the Cutting Edge of Metals Research

A.L. Greer and E. Ma, Guest Editors

## Abstract

Glassy alloys (metallic glasses) are currently the focus of intense research in the international metals community. Setting aside elevated-temperature applications, these amorphous metals have exciting potential for structural applications. When metallic glasses were first widely studied in the 1960s, the alloy compositions then known to be quenchable into the glassy state from the liquid required high cooling rates on the order of  $10^6 \text{ K s}^{-1}$  and were consequently restricted to thin sections. The current interest in metallic glasses has its origin mainly in the increasing range of compositions that can now be cast into glasses at much lower cooling rates, permitting minimum sections of 1 mm to 1 cm or even larger. These bulk metallic glasses (BMGs) are the focus of the articles in this issue of *MRS Bulletin*. Our goal is to illustrate the major materials issues for BMGs, from processing to structures to properties and from the fundamental science of glasses to viable industrial applications. We hope that the articles, in providing a snapshot of a rapidly moving field, show why BMGs are attracting such intense interest and serve to highlight some challenging issues awaiting resolution.

## Introduction

The glassy state is reached when a cooled liquid solidifies without crystallization, which is associated with the phenomena of the glass transition. The focus of this issue of *MRS Bulletin* is on glasses strictly defined in this way, although it should be noted that glasses are but one category of noncrystalline (or amorphous) solids, which in general can be prepared by a variety of methods.<sup>1</sup>

The best known natural glass is obsidian, formed by solidification after volcanic activity. The capability to work obsidian to create extremely sharp cutting edges for knives, spears, and arrowheads made this glass attractive to early humans.

Glassmaking by humans is thought to have begun in Mesopotamia (modern Iraq) around 2500 BCE with the fashioning of beads in imitation of precious stones. In the context of this long history of usage, glasses with nondirectional metallic bonding are *parvenus* among their established cousins. The metallic glasses (also often

referred to as glassy alloys or amorphous alloys) emerged some 50 years ago,<sup>2</sup> defying the expectation that solid metallic states would always be crystalline due to the nature of metallic bonding.

Metallic glasses offer attractive benefits, combining some of the desirable properties of conventional crystalline metals and the formability of conventional oxide glasses. For example, in the absence of the well-defined dislocation defects ubiquitous in crystalline alloys, metallic glasses exhibit room-temperature strength much closer to the theoretical strength of the material than their crystalline counterparts. Meanwhile, near-net-shape processing can be realized by exploiting the viscous flow in the supercooled-liquid regime. Cast glassy alloys exhibit a shiny finish and maintain dimensional accuracy, avoiding shrinkage associated with crystallization. The absence of grain boundaries in glassy alloys contributes to unique combinations of magnetic, electri-

cal, chemical, and tribological properties. In what follows, we highlight the state of the art of research on bulk metallic glasses (BMGs) and introduce the individual articles in this issue, where in-depth discussions can be found on several key issues.

## Processing and Glass-Forming Ability

No pure metals and few metallic alloys are natural glass-formers. A major challenge, therefore, is to obtain glassy alloys in bulk form in a simple operation such as casting. The critical size of BMGs is defined as the maximum possible value of the minimum dimension (such as the diameter of a rod) that permits the sample to be fully glassy. BMGs have indeed been difficult to come by; despite encouraging results on noble-metal-based compositions in the early 1980s,<sup>3</sup> BMG-forming compositions mostly have been discovered only since 1990.<sup>4,5</sup> Systematic research has identified the key thermodynamic and kinetic factors that lead to some alloy compositions with particularly good glass-forming ability (GFA), as analyzed in the article by Busch et al.<sup>6</sup> in this issue. In essence, the alloy melt should have (1) a low entropy and enthalpy and therefore a low thermodynamic driving force for crystallization, and (2) low atomic mobility associated with a viscosity that is high and comparatively weakly temperature-dependent, kinetically suppressing the crystallization. These factors are linked, having their origin in a densely packed liquid structure with pronounced short- and medium-range order.

Guided by such insights, and armed with knowledge of phase equilibria from measurements and calculations, shortcuts to the location of BMG compositions on a phase diagram have been developed in recent experimental searches. These shortcuts have identified a number of new compositions with large GFA, as indicated by the minimum dimension of the largest samples that can be made fully glassy. These developments are summarized in the overview by Li et al. in this issue.<sup>7</sup> As shown in Figure 1, these recently developed BMGs can be cast into rods with diameters in the 1–2.5 cm (1 in.) range and often in systems based on engineering metals such as Mg, Ti, Cu, and Fe. The fast progress on this front during the past few years has several implications. First, it shows that many compositions based on common engineering metals can be processed into BMGs. We are no longer limited to a handful of compositions often containing expensive (such as Pd) or undesirable (such as toxic Be) elements.<sup>5</sup> Second, the high GFAs provide the leeway

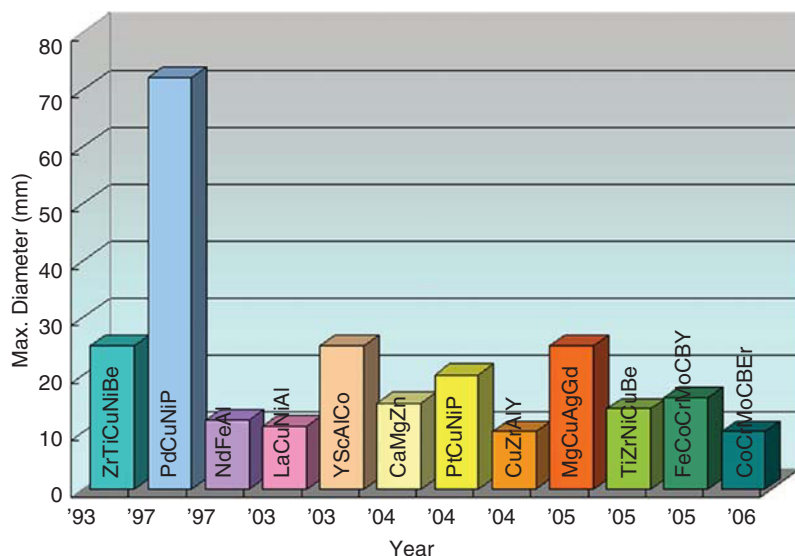


Figure 1. Record-size bulk metallic glasses (BMGs) developed in various alloy systems and the year of their discovery. The first metal (e.g., Zr in ZrTiCuNiBe) is the base element in each alloy; details about the compositions can be found in Reference 7. Alloy systems in which the BMG diameter has not reached 1 cm in copper mold casting are not included.

in processing parameters or composition necessary for reproducibility in industrial mass production. Third, these macroscopic sizes expand the range of possible applications and are in many cases adequate to replace the crystalline counterparts currently in use.<sup>8</sup> Fourth, by moving to nearby compositions on the phase diagram that sacrifice the GFA to some extent, BMG-matrix composites can be designed for a better combination of properties.<sup>7</sup> Ultimately, one would want to predict BMG formation using any chosen element combinations, rather than rationalizing the composition after the fact. We are still far away from such a predictive capability at present. In particular, it is still an unraveled mystery that a small change in composition or a minor addition of alloying elements can drastically change the GFA.

## Structure

Structure determines the properties of materials, and the BMG case is no exception. These glasses exhibit no long-range order, as they are solidified from liquid without reaching the crystalline ground state. But short- to medium-range structural order does develop to a considerable extent, under the given kinetic constraints, as the atoms strive to find comfortable configurations to lower their energy. Short-range order (SRO) develops over the first couple of coordination shells (typically <0.5 nm), beyond which

medium-range order (MRO) may extend to beyond ~1 nm. By SRO and MRO, we mean inherent local structures in the glassy state, not just quenched-in crystallizing phases that are not fully suppressed during solidification. For a monolithic glass, it is the short- and medium-range order that is expected to control its properties, such as the initiation of plastic flow, given the absence of dislocations with defined Burgers vectors.

Understanding how atoms pack in metallic glasses is obviously no trivial task. The structure of metallic liquids, first analyzed by Bernal,<sup>9</sup> was described as dense random packing. For glass-forming alloys in particular, it is clear that dense packing is characteristic. Random packing is certainly incorrect, however, as the densest possible packing of atoms of different sizes can be achieved only with well-developed local order. Structural features of metallic glasses are the focus of the article by Miracle et al. in this issue,<sup>10</sup> where the concept of efficient filling of space is advocated. Importantly, the analysis of dense packing permits rationalization of the good glass-forming compositions. For relatively low solute contents, recent research has established the preference for, and dense packing of, solute-centered quasi-equivalent clusters (Figure 2).<sup>11</sup> An example of a simple binary metallic glass is shown in Figure 2. These structural motifs arise from the strong tendency to form as many bonds as

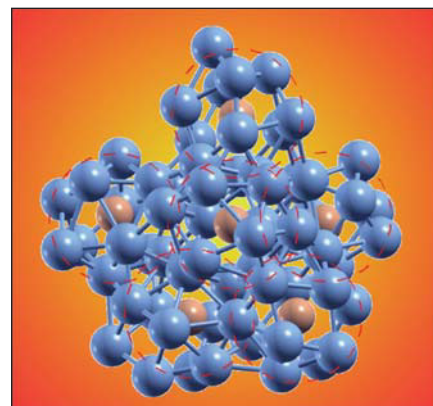


Figure 2. Model of a simple binary metallic glass: interpenetrating quasi-equivalent clusters sharing faces, edges, or vertices in the atomic packing configuration of a Zr-Pt metallic glass. The blue balls represent the solvent Zr atoms centered around Pt solute atoms.<sup>11</sup>

possible between unlike species because of the large negative heat of mixing that is usual in good glass-formers. The cluster size and its type (coordination polyhedron) depend on the relative sizes of the solvent (the majority element) and solute. For example, replacing the Pt solute in Figure 2 with much smaller Be would significantly reduce the number of Zr neighbors that can be accommodated around the solute, and the solute concentration in the alloy would be correspondingly much higher. The clusters overlap via various solvent-atom sharing schemes, leading to medium-range order and dense packing in three-dimensional space.<sup>10,11</sup> Complex structures at more concentrated compositions or in multicomponent systems are yet to be understood.

The dense packing in metallic glasses is of course not unique nor ideal. The cluster packing exhibits a distribution in terms of cluster size, types, and sharing schemes.<sup>11</sup> The concept of free volume,<sup>12</sup> long used to describe the deviation from ideal packing, is critically assessed by Miracle et al. in their article in this issue;<sup>10</sup> recent measurements suggest that microscopic free volume is rather unevenly distributed. The full implications of the structural information acquired so far for atomic diffusion and plastic flow remain to be elaborated. How the structures transform upon changes in thermodynamic and processing variables,<sup>13</sup> or under deformation,<sup>14-16</sup> is only beginning to be investigated. It also seems that our current understanding of metallic glass structures does not yet enable predictions of their stability.

## Mechanical Behavior

From the earliest studies of metallic glasses, it was clear that they have a range of interesting properties. Early work focused on the soft magnetic iron-based glasses, which are highly applicable in the electronics industry. Current work, made possible by the availability of bulk samples, is largely on mechanical properties. As pointed out in the article by Yavari et al. in this issue,<sup>17</sup> these properties are outstandingly good for a variety of structural applications. For example, Figure 3 shows that in comparison with conventional crystalline alloys, BMGs show outstanding strength, elastic strain, and elastic energy storage.<sup>18</sup> Interestingly, BMGs make very good surgical blades, an application harking back to the early use of obsidian glass. But metallic glasses have an Achilles' heel: an almost total lack of tensile ductility at room temperature. This problem, to some extent, hampers application. However, most BMGs are not intrinsically brittle, as are the vast majority of ceramics and glasses, and do possess mechanisms for plastic flow. Metallic glasses just tend to suffer from work-softening and extreme localization of plastic flow into *shear bands*. Yavari et al.<sup>17</sup> have the difficult task of reviewing a fast-moving subject in which great advances are being made in improving plasticity. The guiding principle so far is to facilitate the formation of multiple shear bands so that strains are not concentrated in one or few shear bands that can wreak havoc, not least by evolving into cracks. Large plasticity, in terms of compressive ductility, has been achieved in a number of BMGs,<sup>17</sup> and there are emerging hints that ductility can be achieved even in tension.<sup>19</sup>

In conventional metallurgy, a suitable matrix can be just the basis upon which to optimize microstructure and properties through the dispersion of a second phase. The same is the case with metallic glasses. Many second-phase dispersions are found to be beneficial in improving the strength–plasticity balance. Whereas a range of possibilities is outlined in the articles by Li et al.<sup>7</sup> and by Yavari et al.,<sup>17</sup> the article by Brothers and Dunand in this issue<sup>20</sup> focuses on a gas (or empty space) as the dispersed phase. Porous and foamed metallic glasses as a novel form of BMGs offer nice examples of property tailoring, the benefits of which include lower density, greater plasticity in compression, and greater energy absorption.

Despite recent progress, much remains to be understood about the deformation behavior of BMGs, including the initiation and percolation of plastic flow in the absence of dislocations, the nature and

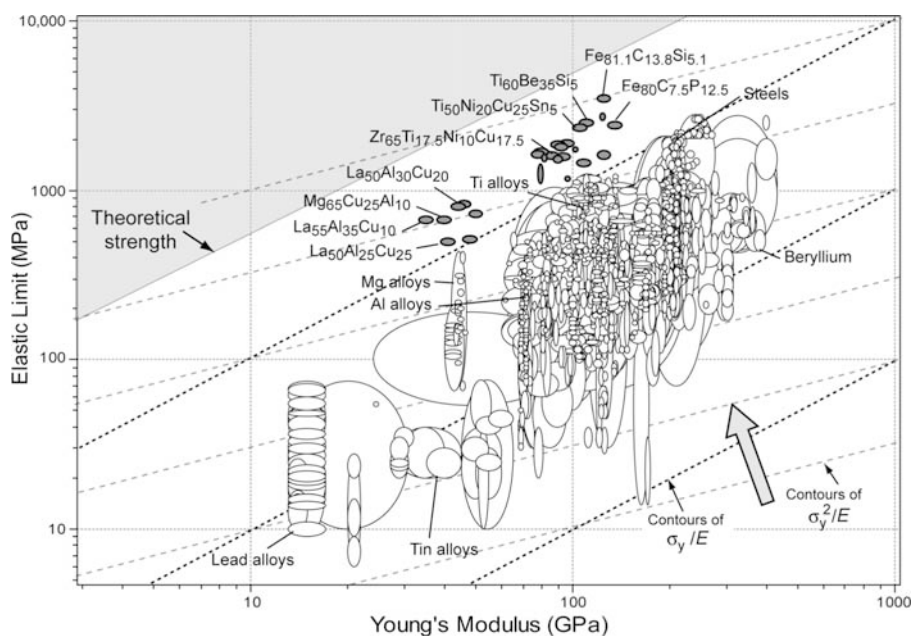


Figure 3. Elastic limit (strength,  $\sigma_y$ ) and Young's modulus  $E$  for bulk metallic glasses (with compositions shown) compared with more than 1500 conventional metals, alloys, and metal-matrix composites. In terms of the  $\sigma_y/E$  (elastic strain) and  $\sigma_y^2/E$  (resilience, or elastic energy storage) contours and of their strength approaching the theoretical limit, BMGs are exceptional.<sup>18</sup>

operation of the shear bands,<sup>21–24</sup> the correlation between the plasticity and the internal structure, the role of nano- to micro-heterogeneities in influencing shear-band nucleation and propagation,<sup>25</sup> and any possible work-hardening mechanisms.

An important step in understanding the rheology of metallic glasses and their parent liquids is described in the article by Johnson et al. in this issue.<sup>26</sup> Such emerging fundamental understanding is a major driver for research on metallic glasses. During the last three decades, several phenomenological models have been proposed to explain flow in metallic glasses, most of which are founded on two hypothetical flow mechanisms. By analogy to granular materials, flow was pictured to occur by deformation-induced dilatation, describable in terms of the creation of microstructural free volume. The latter is responsible for flow localization and consequent softening.<sup>27</sup> In an alternative approach, flow was thought to be accommodated by cooperative shearing of a group of atoms, referred to as the shear transformation zone.<sup>28</sup> Johnson et al. employ a cooperative-shear flow analysis, modeling the shear flow as configurational hopping between inherent states that overcomes an activation barrier in the potential energy landscape.<sup>29,30</sup> They sys-

tematically illustrate, from experimental and simulation results for metallic glass-forming liquids, the key correlations established between viscosity, shear modulus, and potential energy, for a wide temperature range encompassing both Newtonian and non-Newtonian flow regimes.

## Applications

The final article in this issue, by Inoue and Nishiyama,<sup>8</sup> provides an overview of their recent work on applications. They have taken advantage of a number of new BMGs that have recently reached adequate size or possess unique combinations of properties. Many examples in mechanical, chemical, and magnetic applications are explored in their article. In the breadth of their coverage, these authors provide a sense that we are only in the early stages of exploiting the full potential of metallic glasses and there are many opportunities worth pursuing.

Applications in microelectromechanical systems (MEMS) are particularly attractive. This is because metallic glasses offer the complete range of properties desired in such applications, including high strength, extraordinary hardness, superior resistance to wear and corrosion, and the ability to be molded on a very fine scale based on processing in the

supercooled-liquid regime. Meanwhile, the inadequate room-temperature ductility exhibited by large samples becomes irrelevant for small parts, as their size is now below the plastic-zone size of many of the metallic glasses.<sup>18</sup>

As pointed out by Inoue and Nishiyama, the rapid additions of various new BMGs and their composites to the designers' repertoire in recent years greatly facilitate the search for expanded applications. Better GFA and inexpensive compositions based on engineering metals such as Cu, Ni, Fe, Mg, and Al are still being sought after. This effort should continue to be encouraged, especially because a broader impact of BMGs on engineering requires simpler mixtures of elements to lower the cost and facilitate processing, recycling, and commercialization. Mechanical and other property data need to be carefully and systematically documented for metallic glasses to find their niche applications. The continuing development of applications, of course, is a major driver for the research

focus on these materials. It is also our hope that this set of articles can attract the attention of engineers working outside this field to consider BMGs as a new form of material suitable for selection in their innovations.

### Summary and Outlook

As a frontier of metals research, the field of bulk metallic glasses is progressing at a fast pace. At the same time, the unresolved issues in every area of the materials science of these still-novel glasses present major challenges and offer tremendous opportunities for future exploration.

Before closing, we single out one aspect that in our opinion deserves to be highlighted as a key observation and fruitful research direction. Despite many gaps in our knowledge, some of which are highlighted in this collection of articles, it is becoming clear that the properties of metallic glasses are remarkably well correlated with each other. For example, the plastic flow stress scales closely with elastic moduli (Figure 4a).<sup>29</sup> The glass-

transition temperature  $T_g$  provides a suitable basis for normalization of the temperature scale in comparing different glasses, and it too is correlated with elastic moduli<sup>31</sup> and with flow stress (Figure 4b).<sup>32</sup> Furthermore, the elastic moduli are closely correlated with the strong/fragile characteristics of the glass-forming liquid,<sup>31</sup> therefore possibly also with glass-forming ability,<sup>6</sup> and with the toughness/brittleness of the glass (Figure 4c).<sup>33</sup> The close links between the thermodynamic, kinetic, elastic, and plastic properties of metallic glasses are remarkable, and appear to provide predictability for these apparently disordered systems at a level that far exceeds that for their ordered crystalline cousins. A key challenge now is to understand these correlations, which may have common structural origins, and exploit such understanding to develop new glass compositions that combine desirable GFA with mechanical toughness. This would in turn permit more widespread, cost-effective application of these paradigm-shifting materials.

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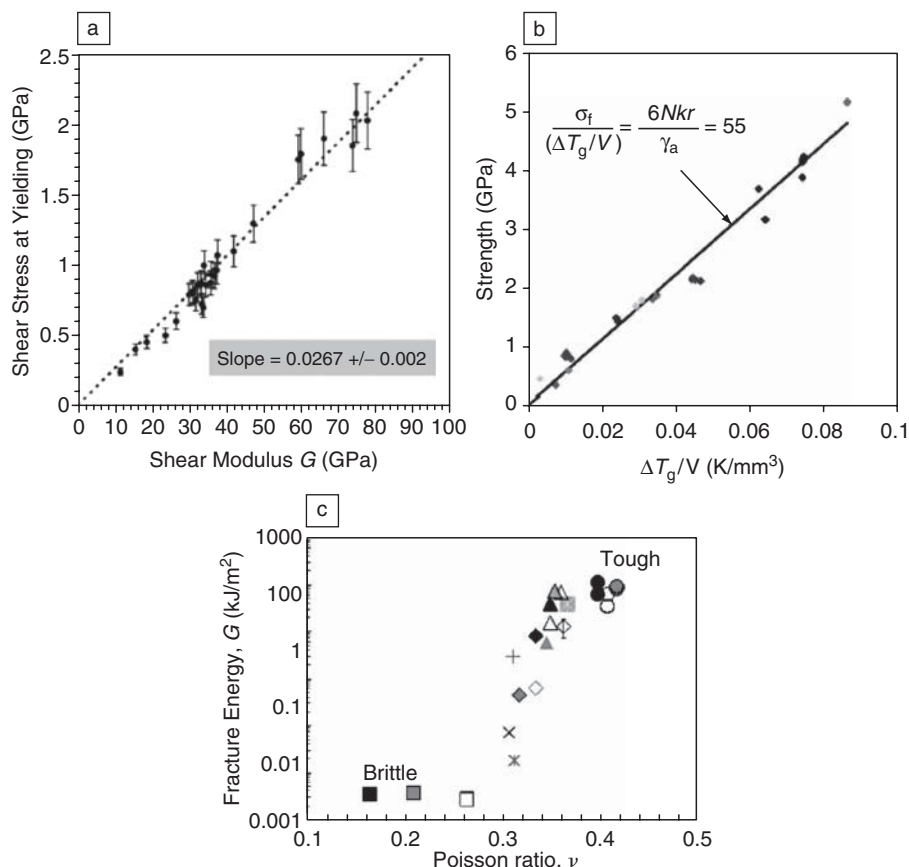


Figure 4. Property correlations for bulk metallic glasses: (a) shear yield strength (50% of the uniaxial strength) scales with shear modulus;<sup>29</sup> (b) uniaxial strength scales with a suitably normalized glass-transition temperature  $T_g$ ;<sup>32</sup> and (c) fracture energy (a measure of toughness) shows a sharp transition with the Poisson ratio  $\nu$ .<sup>33</sup>

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